Origin of $d_{z^2}$ orbital suppression of d-wave superconductive pairs in cuprate

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Compared to Hg-cuprate, the origin that the $d_{z^2}$ orbital suppresses the d-wave superconductive (SC) pairs in La-cuprate is studied based on an effective two-orbital t-J-U model by using the Kotliar–Ruckenstein (KR) slave-boson technique. By analyzing the orbital-dependent electron distribution, it is elaborated that the double occupancy of $d_{z^2}$ orbital, caused by the $d_{z^2}$ orbital mixture, should be responsible for the suppression of the d-wave SC pairs in La-cuprate. When the Coulomb interaction $U$ increases, the ground state hosting the large double occupancy of $d_{z^2}$ orbital in La-cuprate is stabilized by the localization of the carriers due to the Coulomb-blocking instead of reducing the double occupancy by the way of lowering of Coulomb potential energy. Therefore, it could be concluded that the mechanism that the double occupancy destructs against d-wave SC pairs is robust even if the strong Coulomb interaction exists in the La-based compounds.

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1. Introduction

Since the discovery of high temperature superconductivity in copper-based compounds three decades ago [1], the nature and origin of superconductivity has stimulated enormous research interests both experimentally and theoretically. It has become clear that although the physical properties of cuprate compounds in the normal state are still far from the final stage, the SC state is associated with the formation of the Cooper pairs as in the conventional BCS superconductors in spite of the strong evidences for an order parameter exhibiting $d_{z^2}$ symmetry [2–5]. Now, what is remarkable is that the SC transition temperature $T_c$ seems to be material dependent from one family to another, ranging from La$_2$−Sr$_x$Ba$_{2-x}$CuO$_4$ with a maximum $T_c$ of about 40 K to the values as high as $T_c \sim 100$ K in Hg-based compounds. A number of ideas concerning the material dependence of $T_c$ have been discussed: the influence of apical oxygens [6], charge ordering effects [7], band structure variations [8], the type of dopant induced disorders [9], etc. Empirically, the materials with $T_c \sim 100$ K tend to have a round Fermi surface, while the Fermi surface of the La-cuprate is close to a diamond shape. This contradicts the naive expectation of a spin-fluctuation-induced pairing scenario, where a relatively better nesting is more desirable for superconductivity.

Right soon after the discover of superconductivity in the doped cuprate, Anderson [10] suggested that the essential physics of doping cuprate is contained in the t-J model on Cu ion square lattice. This followed from the similar phase behaviors among the different cuprate families hosting high $T_c$ superconductivity: all parent compounds are insulating antiferromagnets with $T_N \sim 300–400$ K, and become SC once doped charge carriers destroy the magnetic order and the Fermi bands are developed. By using the variational Monte Carlo method to the t-J model, the competition between antiferromagnetism and superconductivity was found near half-filling [11], basically consistent with the behavior of cuprates. However, within Anderson’s single-band t-J framework, the materials with the round Fermi surface need to be modeled with large second ($t^* > 0$) and third ($t^* < 0$) neighbor hopping integral, while the low-$T_c$ La-systems have been considered to have small $t'$ and $t^*$ [12]. Indeed, the simple nearest-neighbor (N.N) t-J model cannot be regarded as a comprehensive model when compared quantitatively with the cuprate experiments. The angle resolved photoemission spectroscopy (ARPES) experiments [13] have shown that although the highest energy filled electronic band is well described by the simple t-J model in the direction between the (0, 0) point and the $(\pi, \pi)$ point in the momentum space, but both the experimental data near $(\pi, 0)$ and overall dispersion may be properly accounted by generalizing the t-J model to include the second- and third-nearest neighbor
hopping terms $t'$ and $t''$. The Fermi surface topology observed by ARPES [14] and the change of sign of hall coefficient as a function of doping [15] can best be understood in the presence of $t'$ and $t''$. Moreover, there exist both experimental and theoretical results [16,17] to support that the SC transition temperature is strongly correlated with $t'$ and possible also $t''$.

While the extended $t$-$t'$-$J$ model studies give a tendency consistent with the experiences [18,19], a number of many-body approaches for the Hubbard-like model with realistic values of onsite Coulomb interaction show suppression of superconductivity for large $t' > 0$ and $t'' < 0$ [12]. To resolve this discrepancy, Sakakibara et al. [21] recently suggested a two-orbital Hubbard model which explicitly incorporates the $d_{x^2−y^2}$ orbital on the top of the $d_{xy}$ orbital, and show by the fluctuation-exchange (FLEX) approximation that it is the “single-orbital nature” that favors a higher $T_c$, dominating over the effect of the Fermi surface shape for La-systems. The results are interesting and promising, as they show the existence of various tuning parameters for enhancing the $T_c$. In addition, it has long been known that for the La-cuprate, a band with a strong $d_{x^2−y^2}$ character lies rather close to the Fermi level [22,23]. As for the band filling (number of electrons at per sites), we concentrate on total $n=2.85$, for which the main $d_{x^2−y^2}$ orbital has 0.85.

The interaction term $H_J$ contains a nearest-neighbor antiferromagnetic spin coupling:

$$H_J = \sum_{i,j} J_{ij} S_i^a S_j^a,$$

where $S_{i\alpha} = \sum_\sigma c_{i\alpha\sigma}^\dagger c_{i\alpha\sigma}$ is the spin operator of the electron in the orbital $\alpha$ at the site $i$. It can be believed that the term contains the essential physics for the high $T_c$ superconductivity, since all of the high $T_c$ parent materials show strongly antiferromagnetic spin correlation.

In addition, the multi-orbital local correlation is also added as

$$H_U = \sum_i \left( U \sum_\alpha n_{i\alpha} n_{i\alpha}^\dagger + U' \sum_{\alpha\beta \neq \alpha} n_{i\alpha} n_{i\beta}^\dagger \right) - \frac{J_H}{2} \sum_{\alpha\beta \neq \alpha} \sum_{i,j} c_{i\alpha\sigma}^\dagger c_{i\alpha\sigma}^\dagger c_{j\beta\sigma}^\dagger c_{j\beta\sigma} + J_p \sum_{\alpha\beta \neq \alpha} \sum_{i,j} c_{i\alpha\sigma}^\dagger c_{j\alpha\sigma}^\dagger c_{j\beta\sigma} c_{i\beta\sigma}$$

where the intraorbital $U$, the interorbital $U$ and the Hund coupling $J_H$ (the pair hoping term $J_H$) observe the orbital $SU(2)$ requirement $U = U' + 2J_H$ with $J_p = 0.1U$.

Firstly, one can proceed to decouple the superexchange terms of Eq. (1) by introducing the following order parameters

$$\chi_{i\alpha\beta \sigma \sigma'} = \sum_{j} \langle f_{i\alpha\sigma}^\dagger f_{j\alpha\sigma'} \rangle, \quad \Delta_{i\alpha\beta} = \sum_{\sigma} \eta \langle f_{i\alpha\sigma}^\dagger f_{j\alpha\sigma} \rangle$$

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where $\eta = 1$ for $x$-direction and $-1$ for $y$-direction. Physically, $\chi_{\alpha\beta\sigma\sigma'}$ and $\chi_{i\alpha\beta}$ are the NNN intra-orbital and inter-orbital kinetic average of hopping integrals. $\Delta_{i\alpha\beta}$ and $\Delta_{i\alpha\beta}$ denote the mean-field amplitude of the local NNN intra-orbital and inter-orbital d-wave pairing order parameters. Other symmetric pairs [29,30], although they are possible, will not be considered here. Apparently, in the mean-field treatment, the anti-ferromagnetic exchanges naturally provide for a single d-wave pairing amplitude in the Mott limit, and then the d-wave pairs condensate cooperatively into a d-wave superconductive state upon sufficient doping.

To appropriately treat with the local multi-orbital Coulomb interaction, the Kotilä and Ruckenstein slave-boson technique is employed in the below analysis [31]. By introducing a set of auxiliary bosonic field operators $e, p_{\alpha\sigma}, s_{\alpha\sigma}, d_{\alpha\sigma}$, $h_{i\alpha\sigma}$, $\tau$ to denote each multi-orbital atomic configurations, see Fig. 1, the physical

![Fig. 1. The atomic configurations and the corresponding slave bosons of the two-orbital model.](image-url)
electrons are expressed in terms of the auxiliary bosonic field operators and pseudo-fermionic operators $f_{\text{ion}}$ as

$$c_{\text{ion}} = \tilde{x}_{\text{ion}} f_{\text{ion}}$$

where

$$\tilde{x}_{\text{ion}} = (1 - Q_{\text{ion}}) \tilde{x}_0 Q_{\text{ion}}$$

and

$$z_{\text{ion}} = \tilde{e}_{\text{ion}} p_{\text{ion}}^* p_{\text{ion}} + p_{\text{ion}}^* s_{\text{ion}} + p_{\text{ion}}^* d_{\text{ion}} + s_{\text{ion}}^* d_{\text{ion}} + s_{\text{ion}}^* h_{\text{ion}} + d_{\text{ion}}^* h_{\text{ion}}$$

$$+ d_{\text{ion}}^* h_{\text{ion}} + h_{\text{ion}}^* \tau_i$$

$$Q_{\text{ion}} = p_{\text{ion}}^* p_{\text{ion}} + s_{\text{ion}}^* s_{\text{ion}} + \sum_{\alpha} d_{\text{ion}}^* d_{\text{ion}} + h_{\text{ion}}^* h_{\text{ion}} + \sum_{\alpha} h_{\text{ion}}^* h_{\text{ion}}$$

+ $\tau_i^* \tau_i$.

Formally, the KR slave-boson technique is to utilize the auxiliary bosonic field operators to keep track of the other electrons by measuring the occupancy number in each of possible multi-orbital states which is available for hopping, therefore the hopping term of Eq. (1) is rewritten in the form of the mixed fermionic–bosonic operators as

$$H_t = \sum_{\alpha \beta} \tau_{\alpha \beta}^+ \sum_{\tau \tau'} \tilde{x}_{\alpha \tau} \tilde{x}_{\beta \tau'} f_{\text{ion}}$$

with the set of local constraints

$$1 = \tilde{e}_{\alpha \tau}^\dagger \tilde{e}_{\alpha \tau} + \sum_{\beta \tau} \tau_{\alpha \beta}^+ \sum_{\tau'} \tilde{x}_{\beta \tau'} \tilde{x}_{\alpha \tau}$$

$$0 = \tilde{x}_{\alpha \tau}^\dagger f_{\text{ion}} - Q_{\alpha \tau}$$

to eliminate the unphysical states. In the mean time, the multi-orbital local Coulomb term becomes quadratic in the boson operators

$$H_c = \sum_{\alpha \beta} \tau_{\alpha \beta}^+ \sum_{\tau \tau'} \tilde{x}_{\alpha \tau} \tilde{x}_{\beta \tau'} + \sum_{\alpha \beta} \tau_{\alpha \beta}^+ \sum_{\tau \tau'} \tilde{x}_{\alpha \tau} \tilde{x}_{\beta \tau'}$$

Using the KR slave-boson approach in the mean-field level, all the bosonic field operators are treated as time- and site-independent c-numbers, i.e., their average (or saddle point) or expectation values, since in the low temperature, the slaved bosons occur the Bose–Einstein condensation (BEC), the fluctuations of these boson fields are rather small. In the mean time, the local constraints are released to be as global ones, which can be satisfied by Lagrange multipliers $\lambda$ and $\lambda_{\text{ion}}$. Collecting all the mean-field terms into the Hamiltonian and minimizing the ground state energy, one can obtain the self-consistent equations for the quasiparticles: $d$-wave SC pairing order parameters $\Delta_{\alpha \beta}$, N.N hopping amplitude $x_{\alpha \beta}$, boson field parameter $\tilde{e}_{\alpha \tau}^\dagger$, $p_{\alpha \tau}^*$, $s_{\alpha \tau}$, ..., as well as Lagrange multipliers $\lambda$ and $\lambda_{\text{ion}}$. (Note: the results are tedious and not presented here.)

3. Numerical results and discussions

In this section, we first present the numerical results of two orbital $t$–$J$ model with $U = U' = 0$. In Fig. 2, we show the $d$-wave pairing order parameters $\Delta_{\alpha \beta}$ as a function of the super-exchange constant $J_{\alpha \beta}$. One can clearly note that for both the cases, the dominating pairing correlations $\Delta_{11}$ stem mainly from the $d_{\alpha \beta}^x$ orbital, and are well over the $d_{\alpha \beta}$ orbital pairs $\Delta_{22}$ and the inter-orbital pairs $\Delta_{12}$. In addition, the pairs $\Delta_{22}$ and $\Delta_{12}$ are reduced by the strong Coulomb interaction as shown in Fig. 5(c) and (d), so most probably, they may be not observable. Furthermore, there exists a dramatic difference between the $Hg$- and $La$-cuprate, for example, with $\Delta_{12}$ having a negative value, the $d$-wave superconductive phase is found only for $J > 1.65$ in $La$ systems, contrasting sharply to the rapid and monotinous increase of $\Delta_{11}$ in $Hg$-cuprate. Another more significant result is that the pairing correlation $\Delta_{11}$ for $Hg$-cuprate is much larger than that for $La$ systems for the same $J$ values. That do not only signify a higher $T_c$ for $Hg$-cuprate, but the discrepancy between the extended single band $t$–$t'–J$–$H$ and Hubbard model is simultaneously solved within the present two-band model once the results of Ref. [21] are considered.

To illuminate the reason why the $Hg$- and $La$-cuprate have such a significant different $d$-wave pairing correlation, we investigated the electron distribution of the $d_{\alpha \beta}^x$ and $d_{\alpha \beta}$ orbitals. As shown in Fig. 1, there are four atomic configurations with zero electron, eight atomic configurations with one electron and four atomic configurations with two electrons for the $d_{\alpha \beta}^x$ orbital, therefore the probabilities of empty ($E$, single ($P$) and double ($D$) occupancies on the $d_{\alpha \beta}^x$ orbital should be calculated by the auxiliary bosonic field operators as

$$E = \tilde{e}_{x}^\dagger \tilde{e}_{x} + p_{x1}^\dagger p_{x1} + p_{x2}^\dagger p_{x2} + s_{x}^\dagger s_{x}$$

$$P = p_{x1}^\dagger p_{x1} + p_{x2}^\dagger p_{x2} + d_{x1}^\dagger d_{x1} + d_{x2}^\dagger d_{x2} + d_{x1}^\dagger d_{x2} + d_{x2}^\dagger d_{x1} + h_{x1}^\dagger h_{x1} + h_{x2}^\dagger h_{x2}$$

$$D = s_{x}^\dagger s_{x} + h_{x1}^\dagger h_{x1} + h_{x2}^\dagger h_{x2} + \tau_i^* \tau_i$$

A similar procedure can be applied for the $d_{\alpha \beta}$ orbital as well. The numerical results are displayed in Fig. 3(a) and (b) respectively, where the corresponding occupancy probabilities for $La$-case have been plotted together with that for the $Hg$-cuprate for the sake of comparison. Clearly, the distributions of electron occupancy have a completely different trend for the two studied cases. Firstly, when the atomic configurations with single occupancy are dominant over those with the double and empty occupancies in the main $d_{\alpha \beta}^x$ orbital, the $d_{\alpha \beta}$ orbital of the $Hg$-cuprate is nearly fulfilled with the doubly occupancy $D = 1$, since the realistic energy level difference $\Delta E_{Q}$ was large here. However, in the $La$-cuprate, the distribution of the electron occupancy is nearly the same for the $d_{\alpha \beta}^x$ and $d_{\alpha \beta}$ orbitals, for example, empty ($E$), single ($P$) and double ($D$) occupancies are 0.0885/0.0775, 0.4180/0.5225 and 0.5225/0.5935 for the $d_{\alpha \beta}^x$/$d_{\alpha \beta}$ orbital respectively. At the same time, it could be clearly seen in Fig. 3 that the double occupancy of $d_{\alpha \beta}^x$ orbital is much larger in $La$-cuprate than the one in $Hg$-
cuprate. This is intuitively understandable, since both of the two orbitals are active in La-cuprate due to the strong mixture between the $d_{x^2-y^2}$ and $d_{z^2}$ orbitals around the Brillouin N point [21]. It is well known that in the standard $t$–$J$ model, derived from the large $U$ Hubbard model, the d-wave SC pairs have a robust tendency when the band holds an enhanced single occupancy along with the local constraints which forbid double occupancy at any site. Therefore, it is reasonable to suggest that the enhancement of double occupancy in La-cuprate, which is caused by the $d_{z^2}$ orbital mixture, should be the main reason for the suppression of the d-wave SC pairs.

To further investigate the effects of Coulomb interaction on the electron distribution in the SC state, we calculated the probabilities of the single and double occupancies on the main $d_{x^2-y^2}$ orbital as functions of Coulomb interaction $U$ for the super-change parameter $J_{11} = 3$. In Fig. 4, it is clear that the average double occupancy $D$ and single occupancy $P$ of Hg-cuprate are almost unchanged for small $U < 1.6$, which indicates that the double occupancy is already quite small and the effects of Coulomb interaction are unimportant. And then, $D/P$ decreases/increases gradually as function of $U$, since the energy of the double occupied state is high for large $U$ so that they are energetically unfavorable. However, the results of the two-orbital La-cuprate are completely different from the one of the single-orbital Hg-case, where our numerically obtained $D/P$ show a monotonic and slow decreasing/increasing function of $U$ only for $U < 8.0$, beyond which one obtains the saturating values of $D_0 \sim 0.4$ and $P_0 \sim 0.6$.

Here, the different effects of Coulomb interactions on the electron distribution between Hg- and La-cuprate are related to the orbital-dependent electron dynamics, which can be measured by the bandwidth factors $R_{\alpha\beta}$ in the present KR slave-boson technique. The calculated results are shown in Fig. 5(a) and (b). It is clearly seen that $R_{\alpha\beta}$ of La-cuprate is less orbital-dependent and drastically decreases from 1.0 to less than 0.2 when $U$ increases from 0.0 up to 8.0, beyond which $R_{\alpha\beta}$ are nearly unchanged with increasing $U$. This indicates that as $U$ increases, the electrons are rapidly frozen at per lattice site, lost the ability of transfer, and thus permitted in the ground state with a large double occupancy regardless of the large Coulomb repulsion. On the other hand however, although $R_{\alpha\beta}$ is weightily orbital-dependent, the electrons of Hg-cuprate are still well-itinerant for large but finite $U$, for example, as $U$ increases from 2 to 25, with $R_{x^2z^2}$ being unexchanged, $R_{11}$ decreases from 1 down to 0.60 and $R_{12}$ decreases from 1.0 down to 0.8. Hence, in order to lower the potential energies of system, the double occupied states in Hg-cuprate could be gradually excluded from the ground state. In the other words, as Coulomb correlation $U$ increases, the ground state hosting the large orbital double occupancy in La-cuprate could still be stabilized by the localization of the carriers due to Coulomb-blocking instead of reducing the double occupancy by the way of lowering of Coulomb potential energies.

As has been discussed above, when the local Coulomb correlation partially enforces no double occupancy constraint, the kinetic energies of electrons are at the same time reduced by the bandwidth factor $R_{\alpha\beta}$. As a result, the spin–spin correlation is effectively enhanced, and thus it should favor the d-wave SC pairs. To show this, we display the dependence of d-wave pairs on Coulomb correlation in Fig. 5(c) and (d). In agreement with the $R_{\alpha\beta}$ behaviors, there exits a whole small increase of $\Delta_{11}$ in Hg-cuprate for the range of $U = 0–25$, but in the La-cuprate, the clear increase of $\Delta_{11}$ takes only place for $U < 8$, beyond which $\Delta_{11}$ saturates at $\Delta_{11} \sim 0.13$. Furthermore, we also estimate the ratio of d-wave pairing correlations between Hg- and La-cuprate and bring about $\Delta_{12}/\Delta_{10} \sim 2.5$ for $U = 10$, which strikingly resembles to the ratio of the experimental $T_c$. However, the small d-wave pairing correlation in La-cuprate could not be well understood within the picture that the form of d-wave pairs is determined by the need of reducing kinetic energy [32,33], since the bandwidth factors are in fact very small for the La-cuprate as shown in Fig. 5(b). Therefore, it could be believed that the above mechanism that double occupancy destructs against the d-wave SC pairs is robust even if there exist strong Coulomb interactions in these compounds.

4. Conclusion

In conclusion, using the two-orbital $t$–$U$–$J$ model, we have clearly illustrated the reason why the $d_{z^2}$ orbital suppresses the d-wave SC pairs in the La-cuprate compared to the Hg-cuprate. Our main result is that the orbital-dependent double occupancy, caused by the $d_{z^2}$ orbital mixture, is responsible for the suppression of the d-wave SC pairs in La-cuprate. As the multi-orbital...
Coulomb correlation $U$ increases, the ground states hosting the large double occupancy of $d_{x^2-y^2}$ orbital could be stabilized by the localization of the carriers due to Coulomb-blocking instead of reducing the double occupancy by lowering of Coulomb potential energy, therefore, the mechanism that double occupancy destructs against d-wave SC pairs is robust even if there exists strong Coulomb interaction in these La-based cuprate. Finally, it is rather interesting to notice that the low $T_c$ of recently emerged Fe-based superconductors might be, at least qualitatively, ascribed to their multi-orbital band structure according to our present studies.

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